20

31p

MTP-P&VE-M-62-14
December 19,1962

N63 20215
Code-1

GEORGE C. MARSHALL SPACE FLIGHT CENTER

HUNTSVILLE, ALABAMA

# DEVELOPMENT OF A HIGHLY REFLECTIVE UNFIRED CERAMIC THERMAL INSULATION

by

Vaughn F. Seitzinger

OTS PRICE

XEROX $ 3.60 pl

MICROFILM $ 1.13 mf.

NASA

FOR INTERNAL USE ONLY

GEORGE C. MARSHALL SPACE FLIGHT CENTER

MTP-P&VE-M-62-14

# DEVELOPMENT OF A HIGHLY REFLECTIVE UNFIRED CERAMIC THERMAL INSULATION

By Vaughn F. Seitzinger

## ABSTRACT



A nonflammable, composite, insulation material was developed for protecting the base of the booster stage of the SATURN launch vehicle. The system consists of an unfired, highly reflective, inorganic insulating coating. The coating, designated as M-31, is prepared from fibrous potassium titanate, asbestos fibers, and colloidal silica. It has good thermal shock and erosion resistance and excellent moisture resistance. The material development is described, and selected thermal, optical, and mechanical properties of the coating are reported.

GEORGE C. MARSHALL SPACE FLIGHT CENTER

MTP-P&VE-M-62-14

# DEVELOPMENT OF A HIGHLY REFLECTIVE UNFIRED CERAMIC THERMAL INSULATION

by

Vaughn F. Seitzinger

ENGINEERING MATERIALS BRANCH
PROPULSION AND VEHICLE ENGINEERING DIVISION

TABLE OF CONTENTS

## LIST OF ILLUSTRATIONS



## LIST OF TABLES

GEORGE C. MARSHALL SPACE FLIGHT CENTER

MTP-P&VE-M-62-14

# DEVELOPMENT OF A HIGHLY REFLECTIVE UNFIRED CERAMIC THERMAL INSULATION

by Vaughn F. Seitzinger

## SUMMARY

A nonflammable, composite, insulation material was developed for protecting the base of the booster stage of the SATURN launch vehicle. The system consists of an unfired, highly reflective, inorganic insulating coating. The coating, designated as M-31, is prepared from fibrous potassium titanate, asbestos fibers, and colloidal silica. It has good thermal shock and erosion resistance and excellent moisture resistance. The material development is described, and selected thermal, optical, and mechanical properties of the coating are reported.

## INTRODUCTION

The development of SATURN class launch vehicles presented many major engineering problems. Not the least of these problems was devising a means of protecting the base of the vehicle from the thermal environment to which it was exposed. Base heating in missiles and launch vehicles was not new; however, the degree of heating experienced by SATURN class vehicles was considerably more severe than previous launch vehicles. Specifically, the SATURN C-1, with its cluster of eight 188,000-pound thrust H-1 engines, creates a total thermal heat load of 3600 BTU/$Ft^2$. This heat load is induced by two sources; radiation from the engine plumes, and convection from recirculating exhaust gases. Nominally, 60 - 80 percent of the total heat is from radiation, with the remainder resulting from convection. Although a significant amount of developmental work has been done in the past several years on the development of insulation materials, the majority of such work has been directed toward insulation materials applicable to primarily convective heating environments.

The development of an insulation material for the SATURN base had three primary objectives. First, it was mandatory that the material be capable of providing thermal protection of structural members of the vehicle; maximum temperature limitation affixed to these members was 150°C (300°F). Second, it must be non-burning, since, for a portion of the vehicle flight time, convective cooling of the base resulted if the air scooped into the base was not passed through flames caused by burning of the insulation. Finally, it must be easily applied and air cured since many areas can be protected only after the vehicle is assembled. Oven curing of components assembled on the vehicle cannot be accomplished because of size and because other components in the vehicle cannot stand exposure to the temperatures normally associated with thermosetting materials.

One method of protecting a metal substructure from radiant energy is to apply a coating that will block infrared radiation by diffuse reflectance. Such coatings must be prepared from a material that contains particles having a high refractive index. The size of these particles must adhere to discrete dimensional limitations. Some pigments have these properties; however, it is difficult to prepare thick coatings from them. Therefore, it was necessary to use a fibrous material as the opacifier. After considerable investigation of many materials, fibrous potassium titanate, available commercially under the trade name "Tipersul"*, was determined to have many of the required properties. Therefore, this material was selected as the primary ingredient of the desired insulation material.

---

* "Tipersul" - Trade name of E. I. duPont deNemours Inc., Wilmington 98, Delaware

## INTRODUCTION (CONT)

Binders considered for preparing coatings with low curing temperatures included the sodium silicates, monoaluminum phosphate, "Baymal"* colloidal alumina, and colloidal silica. The sodium silicates lacked refractoriness and had to be cured at undesirable temperatures. Although the monoaluminum phosphate was an excellent bonding agent, it attacked the steel substructure rather vigorously. Colloidal alumina did not provide a suitable bond. Exploratory work indicated that colloidal silica provided a satisfactory bond for "Tipersul" when cured at a temperature as low as 82°C (180°F). Therefore, it was selected as the binder for "Tipersul."

To utilize the capabilities of "Tipersul" as a thermal insulation for metal structures, it was necessary to develop techniques by which a continuous coating of this material could be applied easily and economically. Since a thick coating of the material is required (up to one-half inch in thickness), application by trowelling onto the substructure offered some advantages. Development of this type of coating necessitated investigation of such contributing factors as, (1) the bonding characteristics of various binders, (2) the forms of "Tipersul" most suitable for the desired application, (3) the effect of other inorganic fibers on the strength of the coating, and (4) the type of mechanical device required for adhesion purposes.

Since the primary mechanism of insulation employed by a coating for radiant heating involves the utilization of the optical properties of the materials employed, the development of a coating which would protect these properties during handling and assembly of the components was undertaken. In addition to providing protection of the optical properties of the insulation, the protective coating could not be permitted to adversely affect these properties of the material during or after its removal. Theoretically, a protective coating which would be destroyed quickly by the application of small quantities of heat, and which would leave no residue, would be optimum for this application. Efforts toward the development of such a coating were successful.

## ACKNOWLEDGEMENT

The author gratefully acknowledges the work of A. C. Krupnick in measuring the reflectance and emissivity, C. F. Smith for the specific heat determinations, and A. W. Smock for his assistance in preparation of the data.

* "Baymal" - Trade name of E. I. duPont deNemours Inc., Wilmington 98, Delaware.

## EXPERIMENTAL PROGRAM

### Selection of Materials

1. Opacifier and Filler

Two forms of "Tipersul" fibrous potassium titanate, block and loose fibers, were evaluated as opacifier and filler for the coating. The block form contains approximately 10% "Refrasil"* fibers. The "loose fibers form" does not contain any other inorganic fibers. Table I, taken from reference 1, lists the fiber properties of pure fibrous potassium titanate.

TABLE I

FIBER PROPERTIES OF FIBROUS POTSSSIUM TITANATE

| | |
|---|---|
| Avg. diameter, microns | 1 |
| Length after dispersion, mm. | 0.2 to 0.5 |
| Melting point, $^{o}F$ | 2500 |
| Theoretical density, g/cc<br>or lbs./cu. ft. | 3.6<br>225 |
| Calculated specific heat, BTU/$^{o}F$/lb. | 0.22 |
| Hardness, Mohs | 4.0 |
| Chemical composition, approximate | $K_2 Ti_6 O_{13}$ |

Since the effectiveness of the coating is greatly dependent upon its reflectivity, it must be prepared from raw materials that contain very little contamination. For this reason, the "Tipersul" was heat treated at 538$^{o}C$ (1000$^{o}F$) for 4 hours to remove organic contaminants.

The form of "Tipersul" most suitable for the coating filler was determined by evaluating coatings that contained the block form and/or the loose fibers form. A sol containing 30% colloidal silica was used as the binder for this study. The filler and binder were mixed to the desired consistency and applied, by trowelling to an expanded metal overlay, spot-welded to a steel blank. The specimens were dried by exposing the uncoated side to a bank of radiant heat lamps. Specimens prepared from the block form only,

*Refrasil - Trade name of H. I. Thompson Fiber Glass Co., 1723 Cordova St., Los Angeles 7, California

displayed smaller and fewer cracks and showed less shrinkage than specimens prepared from the loose fibers form. Also, it was determined that the form of "Tipersul" did not appreciably affect the thermal insulating capabilities of the coatings when exposed to a radiant heat flux of 24 BTU/ft$^2$-sec. Consequently, the block form was chosen as the source of "Tipersul" fibrous potassium titanate.

2. Binders

Inorganic binders investigated for preparing coatings with low curing temperatures include sodium silicate, monoaluminum phosphate, "Baymal" colloidal alumina, and colloidal silica. Each binder was used to prepare a set of specimens that contained "Tipersul" block as the filler. All specimens were dried at 93°C (200°F) in an electric oven. After drying, each set of specimens was checked for moisture resistance by immersing them for 3 hours in boiling tap water having a pH of 7.3. The only specimens that did not disintegrate during the moisture resistance test were those bonded with colloidal silica. Also, the coating prepared with colloidal silica had a hard, dense outer layer formed by silica colloids that had migrated toward the surface during the drying process. The outer layer lends strength and rigidity to the coating. The inner portion of the coating is much lighter and serves as an excellent thermal insulator. For these reasons, colloidal silica was considered the most attractive bonding agent for "Tipersul"-filled thermal insulations. In this investigation, "Ludox HS"*, a colloidal silica sol that contains 30% solids, was used as the binder.

3. Inorganic Fiber Additives

Because the block "Tipersul"-filled coatings cracked excessively during drying, it was necessary to use another inorganic fiber with larger and longer filaments. This additive was designed to decrease the shrinkage, thus decreasing the cracking. Both asbestos and refractory glass fibers were considered for this addition. Both fibers were effective in decreasing the shrinkage; however, the refractory glass fibers increased the density of the finished material considerably. Therefore, a long fiber asbestos** was chosen as the additive. To insure that the asbestos fibers could be worked through the openings of the expanded metal, they were cut to approximately ½ inch lengths. After determining the effects of asbestos on the strength and infrared reflection of the coating (both are discussed in the results and discussion section), it was decided to use a 10% asbestos fiber addition.

---

* "Ludox HS" - Trade name of E. I. duPont deNemours Inc., Wilmington 98, Delaware.

** Asbestos - Number 1 Chrysotile AAAA grade with a maximum iron content of 1.3 percent by weight reported as ferrous oxide (FeO)- Asbestos Corporation of America, 31 North Ave., Garwood, New Jersey.

## Mixing

The cut asbestos fibers and "Tipersul" block were blended into a uniform mixture with a Patterson-Kelly, Double Cone Mixer. The coating was prepared by mixing the batch materials in the bowl of a Hobart Kitchen Aid Mixer. The amount of colloidal silica sol added was sufficient to give a trowellable consistency. The final composition, designated M-31 thermal insulation, is listed in Table II.

TABLE II

BATCH COMPOSITION OF M-31 THERMAL INSULATION

| Material | Parts by Weight |
|---|---|
| "Tipersul" Block | 90 |
| Cut Asbestos | 10 |
| "Ludox HS" | 420 |

## Application

To apply M-31 insulation to metal airframes, it is necessary to use some type of mechanical device for bonding purposes. A recent report by Sklarew, Hauch, and Levy (Ref. 1), on the development of insulating coatings for ram-jet applications, indicated that a metal mesh reinforcement improved the structural stability of a ceramic insulating layer. After experimenting with various reinforcements, these investigators concluded that an expanded metal gave the best performance. It provides an excellent mechanical interlock because its sides are not vertical but are set at an angle.

For this investigation, the expanded metal was fabricated from 26-gage SAE 1015 steel, having diamond shaped openings approximately ½ inch across the longest dimension and ¼ inch across the shortest. The overall thickness of the material was approximately 0.080 inch, and it weighed about 0.27 pound per square foot. The expanded metal was attached to its steel substrate by spot-welding. To assure the best possible spot-welds, both the expanded metal and its steel substrate were cleaned with an organic solvent before the spot-welding operation.

The M-31 was applied to its substructure by trowelling and rolling. Care was exercised to insure that a coating of uniform thickness was applied and that it was interlocked in the expanded metal overlay.

Drying

M-31 thermal insulation can be cured by simply removing the water. This can best be accomplished by uniformly heating the coated panel. For this investigation, a bank of infrared heat lamps was used for drying the M-31 as follows: (1) the surface temperature of the uncoated side was raised from ambient to 49°C (120°F) in 1 hour and held at this temperature for 4 hours; (2) the surface temperature was raised from 49°C (120°F) to 82°C (180°F) over a period of 10 hours and held at this temperature for 2 hours. All temperatures were controlled within ±6°C (10°F). A typical drying curve and the related moisture content of the coating are shown in Figure 1.

The few drying cracks that may form can be filled easily with a grout compounded from 1 part, by weight, of heat treated [538°C (1000°F) for 4 hours] fibrous potassium titanate loose fibers and 6 parts, by weight, of "Ludox HS" colloidal silica. This grout has been designated M-31-R.

Protective Coating

Because the effectiveness of M-31 insulation depends greatly upon its optical properties, a protective coating was required to keep its outside surface from being contaminated during assembly and other handling operations. Two types of protective coatings were given consideration. The first type consisted of a plastic strippable coating, applied over the M-31, which could be removed prior to flight. The second type was one that could be readily consumed by the heat absorbed from the engine exhaust gases without leaving any residue that would offset the optical properties of the surface of the M-31. Since it was not desirable to use a coating that had to be stripped prior to flight, the second type of coating was given primary emphasis. After considerable study, it was decided to use nitrocellulose* for preparing a coating that would burn off quickly. The particular nitrocellulose used contained 11.8 - 12.2% nitrogen. The nitrocellulose was dissolved in sufficient methyl-ethyl-ketone (MEK) to give a zahn #3 viscosity of 22 plus or minus 2 seconds. The solution was then colored with a dye consisting of a saturated solution of Janus Green** (Diazine Green) in ethyl alcohol. The purpose of the coloring agent is to increase the heat absorptivity of the protective coating, thus accelerating burn-off.

The nitrocellulose coating was applied by brushing. Three applications, each approximately 1.5 mils dry thickness, were required. Each application was dried tack-free before the next one was applied. The protective coating has been designated M-31 C.

---

* Nitrocellulose - Type HB14E manufactured by E. I. duPont deNemours and Co., Carney's Point Works, Penns Grove, New Jersey.

** Janus Green - National Aniline & Chemical Co., Inc., New York, N. Y.

Although this protective coating will burn off quickly in the presence of heat, the flame readily extinguishes itself when the heat is withdrawn.

## RESULTS AND DISCUSSION

### Engineering Properties

To evaluate the capabilities of M-31 thermal insulation, it was necessary to make several property determinations. These determinations were made on the M-31 ceramic material only unless otherwise noted. Properties of engineering interest that were determined are as follows: (1) bulk density and water absorption, (2) hardness, (3) moisture effect, (4) refractoriness, (5) thermal-shock resistance, (6) reflectance (7) emissivity, (8) specific heat, (9) thermal conductivity, and (10) mechanical strength. Also, the effects of vibration and flexure, and radiant heating were determined.

1. Bulk Density and Water Absorption

Bulk density and water absorption were determined by means of ASTM standard procedures for refractory materials (Ref. 3). These determinations were made on specimens of both ¼ inch and ½ inch thicknesses of M-31 material.

Results of these determinations showed that there was no appreciable difference in either the bulk density or water absorption of the two thicknesses of M-31. The average values for the bulk density were 0.75- 0.80 grams per cubic centimeter (46.8 - 49.9 pounds per cubic foot), and the water absorption 75-77 percent.

As reported in a previous section, silica colloids migrate toward the surface of M-31 during the drying process. The result of this phenomenon is a density gradient through the thickness of the coating. To illustrate this density gradient, the densities of selected layers of a specimen approximately ¼ inch thick were determined as follows: (1) the density of the ¼ inch thick specimen was determined, (2) a thin layer of the outer surface of the specimen was removed by grinding, and the density of the removed material calculated, and (3) step (2) was repeated several times until only a thin layer of the specimen remained. The results are illustrated in Figure 2.

The illustration shows that the density of the specimen is highest at the front face, lowest near the middle portion, and intermediate near the back face. This indicated that either some of the silica colloids did not migrate toward the front face during drying, but remained in situ near the back face of the specimen, or the migration occurred in both directions.

The high density of the outer surface causes it to be sufficiently hard to resist ablation produced by high velocity exhaust gases.

2. Hardness

Hardness measurements were made with a model 4JR Rockwell Superficial Hardness Tester on the outer layer of M-31 coatings that had been completely dried. The measurements were made with a 60-kilogram load using a 1/8 inch diameter steel ball (H-scale). The hardness of the outer layer, which was approximately 0.060 inch thick, was found to be about H98.

The outer surface of fully dried M-31 insulation was determined to have a Mohs hardness of about 6.

3. Moisture Effect

The moisture effect on dried M-31 insulation was determined by a boiling-water test. The thicknesses of four specimens of the coating were measured to the nearest 0.0001 inch at selected spots. The specimens were then immersed for 3 hours in boiling tap water having a pH of 7.3. Care was exercised to insure that the specimens did not come in contact with the bottom of the water container. After boiling, the specimens were dried in an electric drying oven. The thicknesses were measured again at the same spots as before. The average thickness loss was determined to be less than 0.12 percent. This indicates that boiling water does not appreciably affect the stability of the material.

4. Refractoriness

An indication of the refractoriness of M-31 was obtained by preparing the coating into bar specimens and heating these bars, which were supported at only one end, in an electric furnace. The furnace temperature was raised at approximately 111°C (200°F) per hour. No change was noted in the position of the bars up to 871°C (1600°F). At this temperature the bars had softened sufficiently to cause them to bend slightly. From this point on, the bars were removed from the furnace after each 56°C (100°F) temperature rise and examined visually. No other change in the refractoriness of the coating was noted until the temperature reached 1204°C (2200°F). At this temperature it was noticed that some melting of the inner part of the coating had occurred. There was no melting of the outer layer of the material up to a temperature of 1316°C (2400°F). The test was terminated at this point.

5. Thermal-Shock Resistance

The thermal-shock resistance of both expanded metal reinforced and unreinforced M-31 material, not bonded to a substrate, was determined. The test consisted of inserting specimens (0.25" X 3" X 3") in a hot furnace, allowing them to remain until they reached the furnace temperature, removing and quenching the specimens in water. A fresh set of three samples

of both the reinforced and unreinforced M-31 was subjected to this treatment at each temperature. The temperatures used were 427° (800°), 538° (1000°), 649° (1200°), 760° (1400°), 871° (1600°), and 982°C (1800°F).

All specimens withstood the cold shock when quenched from 871°C (1600°F) and below. Both the reinforced and unreinforced specimens failed when quenched from 982°C (1800°F). At this temperature, the unreinforced coating cracked badly. The reinforced specimens also cracked badly; however, the coating did not separate or pull away from the expanded metal reinforcement.

6. Reflectance

A Perkin-Elmer Model 112 double-pass, single-beam spectrophotometer equipped with a special integrating sphere was used to measure the absolute spectral reflectance in the 0.30-3.0 micron wavelength range. A Hanovia continuous spectrum hydrogen arc source was used in the 0.30 - 0.34 micron wavelength range; a tungsten lamp was used in the 0.34 - 0.85 micron range; and a Globar source was used in the 0.90 - 3.0 micron range. A standard IP-28 photomultiplier tube was used as the detector in the 0.30 - 0.85 micron range, and a lead sulfide cell was used in the 0.90 - 3.0 micron range.

The absolute spectral reflectance values are given in Figure 3.

7. Emissivity

A modified Perkin - Elmer Model 21 Infrared Spectrophotometer was used to measure the spectral emissivity in the wavelength region between 3 and 14 microns. The emissivity furnace is mounted upon the spectrophotometer in such a manner that both the furnace (black body radiator) and the sample can be monitored simultaneously by the spectrophotometer. An Inconel K Furnace heated to 800°C (1472°F) maintained by three individually controlled resistance heaters, is used to provide thermal radiation. This furnace has a roof inclined at 7° with an opening so that the sample forms an integral part of the furnace. The multiple reflection arising from the furnace walls and sample are brought to focus upon the monochromator entrance parts by a series of front-surfaced aluminum mirrors similar to the method used by Reid and McAlister (Ref. 4). Because of the relation:

$$E = 1-R$$

where

E = Emissivity
R = Reflectance,

the spectral emissivity of a material can be determined by a practical and highly accurate method.

The samples are placed in a water cooled jacket with water impinging directly on the back face of the sample to control the temperature. Variations in the sample temperature can cause serious errors in the emissivity data obtained; therefore, sample temperatures must be controlled within 1°C (2°F).

The spectral emissivity values are given in Figure 4.

8. Specific Heat

Specific heat determinations were made by the method of mixtures, using a water calorimeter. The calorimeter was calibrated using pure electrolytic copper. The specific heat of M-31 in the 25-85°C (77-185°F) temperature range was determined to be 0.31 BTU/lb/°F.

9. Thermal Conductivity

The thermal conductivity measurements of M-31 were made by the Midwest Research Institute, Kansas City, Missouri. The results are given in Figure 5.

10. Mechanical Strength

For convenience of testing, a transverse (flexure) test was used for determining the mechanical strength of M-31. The measurements were made on an Instron Model TT-B tensile testing machine. The test specimens were 6 inches long, 1 inch wide, and ½ inch thick. They were end supported on round bars, having a 7/16 inch radius, and spaced 5 inches apart. The load was applied at the center point perpendicular to the front side of the M-31 specimens, unless otherwise noted. The crosshead (loading) speed was 0.05 inch per minute. The depth and breadth of the specimens were taken at the break. The transverse strength (modulus of rupture) was calculated by the formula:

$$M = \frac{3Pl}{2bd^2}$$

where
M = Modulus of Rupture in Pounds Per Square Inch
P = The Breaking Load in Pounds
l = Distance Between Supports in Inches
B = Width of Specimen in Inches
d = Depth of Specimen in Inches

The effect of water on the mechanical strength of M-31 was determined by making strength measurements on the material after it had been subjected to one of the following treatments: (1) soaked in 75°F water for 100 hours, (2) soaked in 75°F water for 100 hours and dried, and (3) immersed in boiling water for 3 hours and dried.

To determine the effect of the dense outer layer, strength measurements were made by applying the load from the back side of the material.

The results of the strength determinations are given in Table III.

TABLE III

MODULUS OF RUPTURE OF M-31

| MATERIAL AND CONDITIONS | MODULUS OF RUPTURE PSI |
|---|---|
| M-31, As Prepared | 475 |
| M-31, Soaked in Water for 100 Hours-Tested Wet | 370 |
| M-31, Soaked in Water for 100 Hours and Dried | 480 |
| M-31, Immersed in Boiling Water for 3 Hours and Dried | 485 |
| M-31, Load Applied From Back Side | 1,075 |

Table III indicates that M-31 in the wet condition is approximately 22 percent weaker than the as-prepared material. However, water seems to have no effect on the strength of the material provided it is dried before being stressed. Also, Table III shows that the material is approximately twice as strong when loaded from the back side. This result was expected since this type of loading subjects the dense outer layer of the material to the greatest stress.

Young's modulus (E) was also determined for M-31. The data used for calculating Young's modulus were obtained simultaneously with the data used for calculating the modulus of rupture. The results are given in Table IV.

TABLE IV

YOUNG'S MODULUS (E) OF M-31

| MATERIAL AND CONDITIONS | YOUNG'S MODULUS (E) PSI |
|---|---|
| M-31, As Prepared | $2.5 \times 10^5$ |
| M-31, Soaked in Water for 100 Hours-Tested Wet | $2.0 \times 10^5$ |
| M-31, Soaked in Water for 100 Hours and Dried | $2.5 \times 10^5$ |
| M-31, Immersed in Boiling Water for 3 Hours and Dried | $2.7 \times 10^5$ |
| M-31, Load Applied From Back Side | $2.6 \times 10^5$ |

VIBRATION AND FLEXURE

A preliminary M-31 type coating was applied to a steel heat shield panel (30" X 54") and tested on SA-T. It contained only "Tipersul" as the filler. The panel was first overlaid with an expanded metal attached by spot-welding on approximately 6 inch centers. A 0.280" thick coating was applied above the base metal. This coating failed by separating from the expanded metal, because it did not have sufficient strength to withstand the vibration and flexure encountered during testing. Failure of this type had not been experienced in the laboratory even though vibration was included in the test conditions. A program was initiated to develop a coating with greater strength. Since asbestos was effective in decreasing shrinkage, it was decided to determine its effect on the strength of such a coating. Specimens containing 0, 5, and 10 percent asbestos fibers were prepared and applied to an expanded metal spot-welded to steel blanks. Relatively thin steel blanks (0.038" X 6" X 11") were selected so that the specimens would flex in the vibration test.

A program of vibration and flexure was initiated in which all specimens were tested to destruction with increasing "g" loadings. Specimens that did not contain asbestos fibers sometimes failed within the coating itself. Specimens containing either 5 or 10 percent asbestos always failed at the spot-welds attaching the expanded metal to the steel blanks. Failure of specimens containing 10 percent asbestos (M-31) generally occurred at 35 to 50 g (1/8 to ¼ inch deflection within the 11-inch length of the specimen). This is approximately 25 percent higher than the g load required to cause failure of specimens containing only 5 percent asbestos. One specimen of M-31 withstood 72 g for 15 seconds after 4

minutes at an average of 40g vibration. Of specimens tested after soaking in water for 170 hours, failure generally occurred at 40g; however, one specimen withstood almost 90g for 7 seconds after 2 minutes at 40g. Generally the deflection of the wet samples was greater than that of the dry specimens at the same frequency because of their greater weight. Failure was still in the spot-welds, not in the material.

## RADIANT HEATING

The radiant heating test apparatus (Figure 6) consisted of: (1) a lampholder; (2) a gold-electroplated-on-copper lamp reflector--water cooled; (3) a vibrating device; and (4) necessary accessory equipment. The lampholder accommodates seventeen 1000 watt, 220 volt, quartz-tube infrared lamps. Unless otherwise noted, all specimens were tested at a heat flux of 24 plus or minus 1.2 BTU/ft$^2$-sec while being vibrated at 11g.

To show the effects of various "Tipersul"-base coatings as reflectors of radiant energy, the following samples were prepared and tested: (1) a mild steel blank (0.080" X 6" X 11") without a coating, (2) a coating of 50 percent loose fibers "Tipersul" and 50 percent block "Tipersul," (3) a coating of 100 percent block "Tipersul," (4) a coating of 95 percent block "Tipersul" and 5 percent asbestos fibers, and (5) a coating of 90 percent block "Tipersul" and 10 percent asbestos fibers, M-31. All samples were prepared in the same manner as described earlier for M-31, and the substrates were expanded metal overlays spot-welded to mild steel blanks that were 0.080 inch thick, 6 inches wide, and 11 inches long. All coatings were approximately 0.320 inch thick. A thermocouple was spark-welded to the back face of each specimen. The effect of composition on the performance of "Tipersul" as a reflector of radiant energy is illustrated in Figure 7 where it is shown that the form of "Tipersul" has very little effect on the back face temperature rise. However, additions of asbestos fibers definitely decrease the effectiveness of "Tipersul" as a radiant heat reflector. The illustration also shows the effectiveness of M-31 as a radiant heat reflector; the temperature rise for the first 8 seconds of test was about 0.6°C (1°F) for the M-31 coating compared to about 222°C (400°F) for the mild steel blank.

Subsequent to these tests, a limited number of tests were made on the M-31 material at a pressure of approximately 2 mm Hg. It was determined that at this pressure, and during exposure to heating rates greater than 40 BTU/ft$^2$-sec, an apparent chemical reaction occurred in the material causing a significant reduction in optical properties. Although not proven at this writing, it is theorized that the potassium titanate undergoes a chemical change with a resultant change in optical properties. As a result of these tests, it is not recommended that the M-31 material be considered for applications when the combined pressure-heating rate

environment equals or exceeds those listed above. Since it is believed that pressure, heating rate (temperature) and time are variables which affect the chemical reaction, further work is required to define the limitations of the material under reduced pressure.

The effect of water on the thermal performance of M-31 was determined. Specimens were prepared by applying M-31 to expanded metal overlays spot-welded to blanks of mild steel that were 0.038 inch thick, 6 inches wide, and 11 inches long. Some of the specimens were soaked in water for 170 hours which increased their weight approximately 75 percent. Both the wet and dry samples were exposed to radiant heating. The results obtained are given in Figure 8,which shows that after 40 seconds of exposure, the temperature of the specimens containing water rose rapidly until it reached 100°C (212°F), the boiling temperature of water. At this point, the temperature remained constant for the duration of the test, thus, indicating that all of the water had not been removed. The rapid temperature change of the specimens containing water is attributed to the high thermal conductivity of water. After 145 seconds of exposure the temperature rise of the wet M-31 was only about 55 percent of that of the dry M-31. This indicates that water actually enhanced the effectiveness of the insulation at one atmosphere pressure when exposed to a radiant heat flux of 24 BTU/ft$^2$-sec.

The effect of protective coatings on the temperature rise of M-31 was studied. Specimens used for this evaluation were 4 inches wide and 6 inches long. The insulation (approximately 0.340 inch thick) was applied to an expanded metal spot-welded to a steel blank that was 0.080 inch thick. A thermocouple was spark-welded to the back face of each specimen. The protective coatings were applied by brushing. Three coats of either the nitrocellulose or the BFC Nr. 3253 Liquid Envelope* strippable coating were applied to each sample. The results are shown in Figure 9.

Both the nitrocellulose and BFC Nr. 3253 started flaming at about 5 seconds after exposure and burned for approximately 2 seconds. At this point, the coatings charred leaving a carbon residue on the surface of the insulation. At about 18 seconds after exposure, the carbon residue on the samples coated with nitrocellulose was completely burned off. It took approximately 30 seconds to burn off the carbon residue left by the BFC Nr. 3253 coating. Because the carbon caused the specimens to absorb heat, the temperature rise was highest for those specimens on which the carbon remained the longest. Therefore, nitrocellulose was more attractive as a protective coating than the BFC Nr. 3253 strippable coating.

Efforts to decrease the burn-off time by adding selected oxidizers to the nitrocellulose protective coating showed some promise; however, time did not permit a complete investigation.

---

* Better Finishes and Coatings, Inc., 268-276 Doremus Avenue, Newark 5, New Jersey

## CONCLUSIONS

A new type of ceramic coating has been developed in which "Tipersul" inorganic fibers are bonded with a colloidal silica sol. The coating does not require a high curing temperature; drying at 82°C (180°F) imparts stability to the material and provides satisfactory moisture resistance. The coating, after drying, is a low density material (46-50 lbs./cu. ft.) that has a hard, dense outer layer. The inner portion is much lighter and is an excellent thermal insulator. The thermal conductivity is between 0.85 - 1.20 BTU/ft$^2$-hr °F/in. in the 93 - 760°C (200 - 1400°F) temperature range. The specific heat is approximately 0.31 BTU/lb./°F in the 25 - 85°C (77 - 185°F) temperature range.

A 0.320 inch thickness of the coating was not affected when exposed to a radiant heat flux of 24 BTU/ft$^2$-sec for 145 seconds, and the back face temperature rise of the specimen was only 109°C (228°F). Water enhanced the effectiveness of the insulation when exposed to a radiant heat flux of 24 BTU/ft$^2$-sec., without increasing the susceptibility of the coating to thermal shock.

Additions of asbestos fibers decreased the shrinkage of the coating and increased the strength. Specimens containing asbestos fibers withstood high frequency vibration up to 72 g's before failing. Failure always occurred at the spot-welds attaching the expanded metal to the steel blanks, and not within the coating.

The greatest promise for this new coating is probably its use as an insulator for protecting airframes against radiant heat. Its absolute spectral reflectance lies between 80-90 percent in the 0.40 - 1.90 micron wavelength range, and its spectral emissivity is between 0.60 - 0.66 at 321°K (578°R) in the 5 - 13 micron wavelength range. Although it is recognized that colloidal silica bonded "Tipersul" has limitations, with respect to reduced pressure applications, it is concluded that by proper application this material can be utilized where many other materials might not withstand environmental conditions.

## REFERENCES

1. Technical Information Bulletin Nr. 3 on "Tipersul", E. I. duPont de Nemours Inc., Pigments Department, Wilmington 98, Delaware.

2. Sklarew, S., Hauck, C. A., and Levy, A. V.: Development and Evaluation of Insulating Type Ceramic Coatings, Part 1, Development and Small Scale Testing. WADC TR 57-5777, Aug., 1957.

3. Apparent Porosity, Water Absorption, Apparent Specific Gravity, and Bulk Density of Burned Refractory Brick. ASTM Designation: C20-46, Book of Standards, Part 5, pp 374-379, 1958.

4. Reid, Charles D., and McAlister, D. D.: Measurement of Spectral Emissivity from 2 to 15 Microns. Journal of the Optical Society of America, Vol. 49, No. 1, Jan., 1959, PP 78-82.

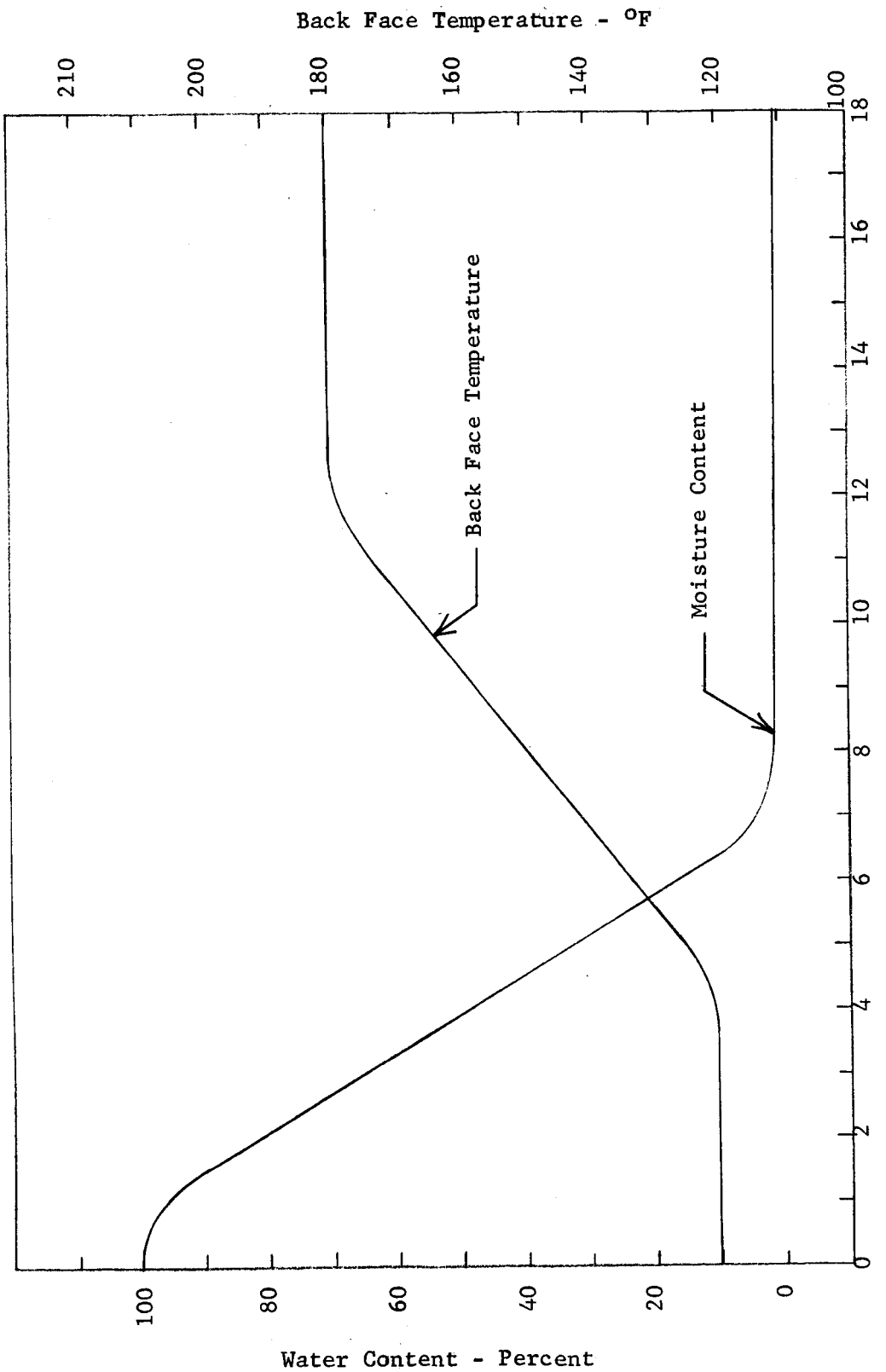


Figure 1 - Drying Curve for M-31

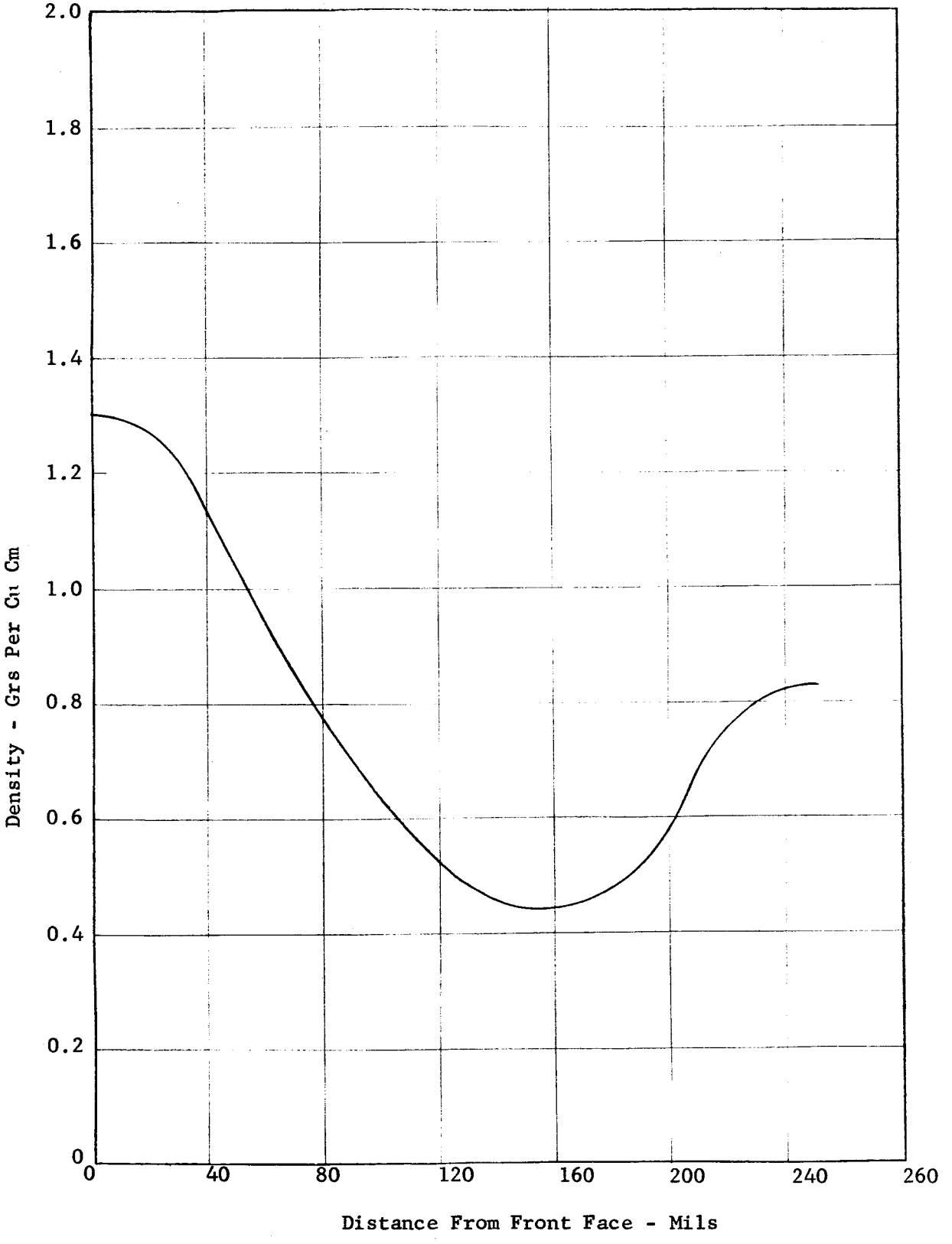


Figure 2 - Density Gradient Through a ¼ Inch Thick Specimen of M-31

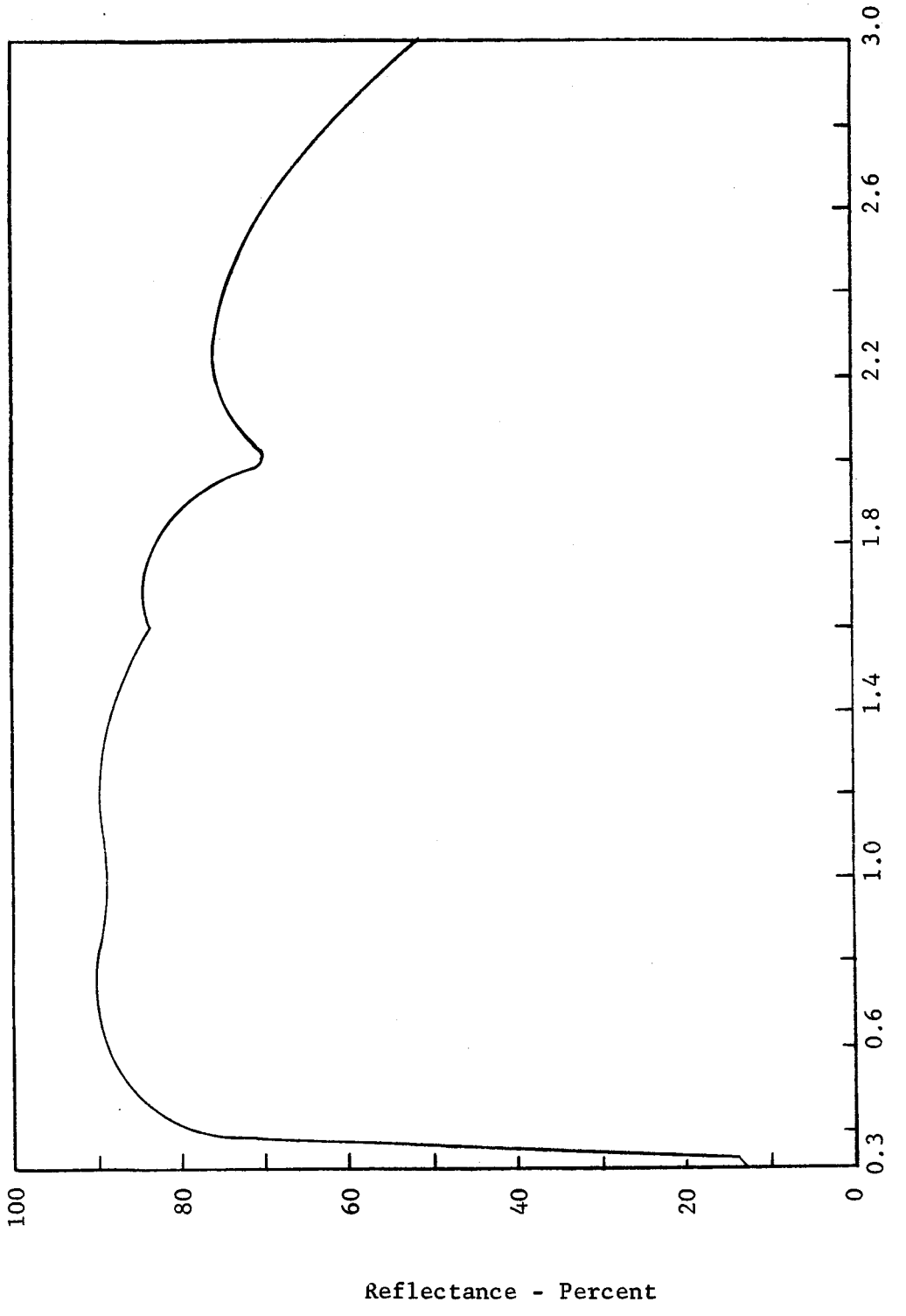


Figure 3 - Absolute Spectral Reflectance of M-31

MTP-P&VE-M-62-14

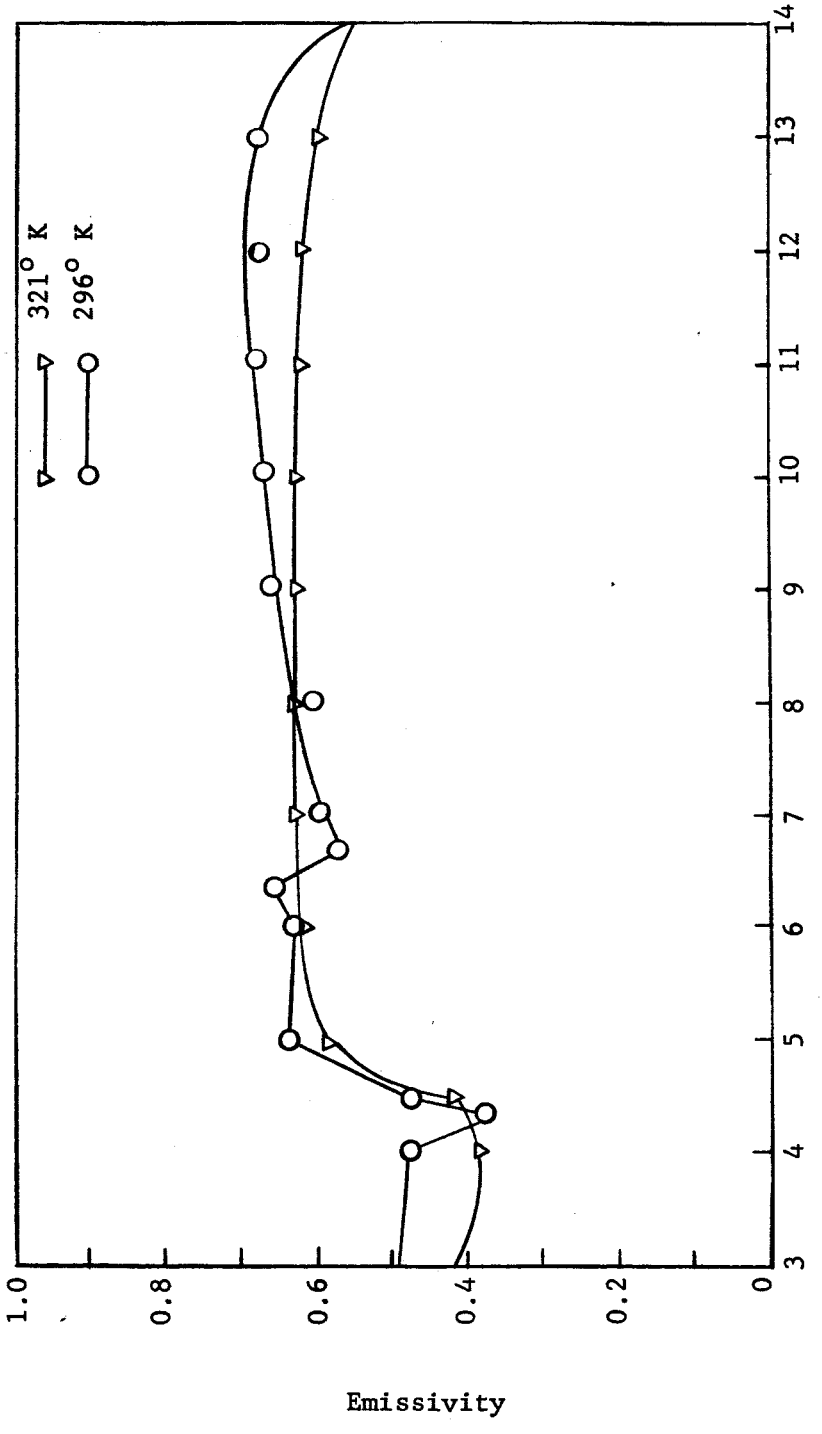


Figure 4 - Spectral Emissivity of M-31

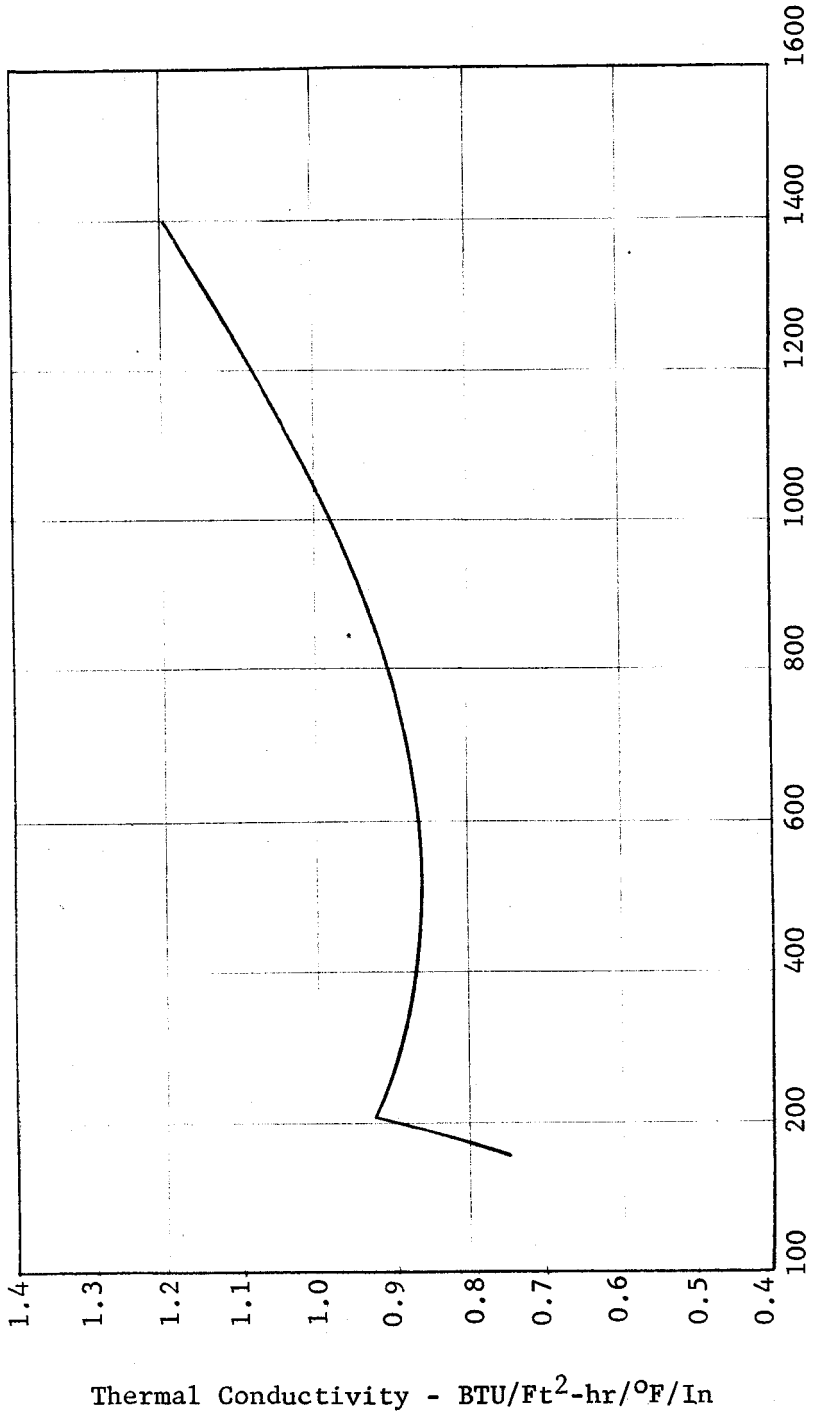


Figure 5 - Thermal Conductivity of M-31

MTP-P&VE-M-62-14

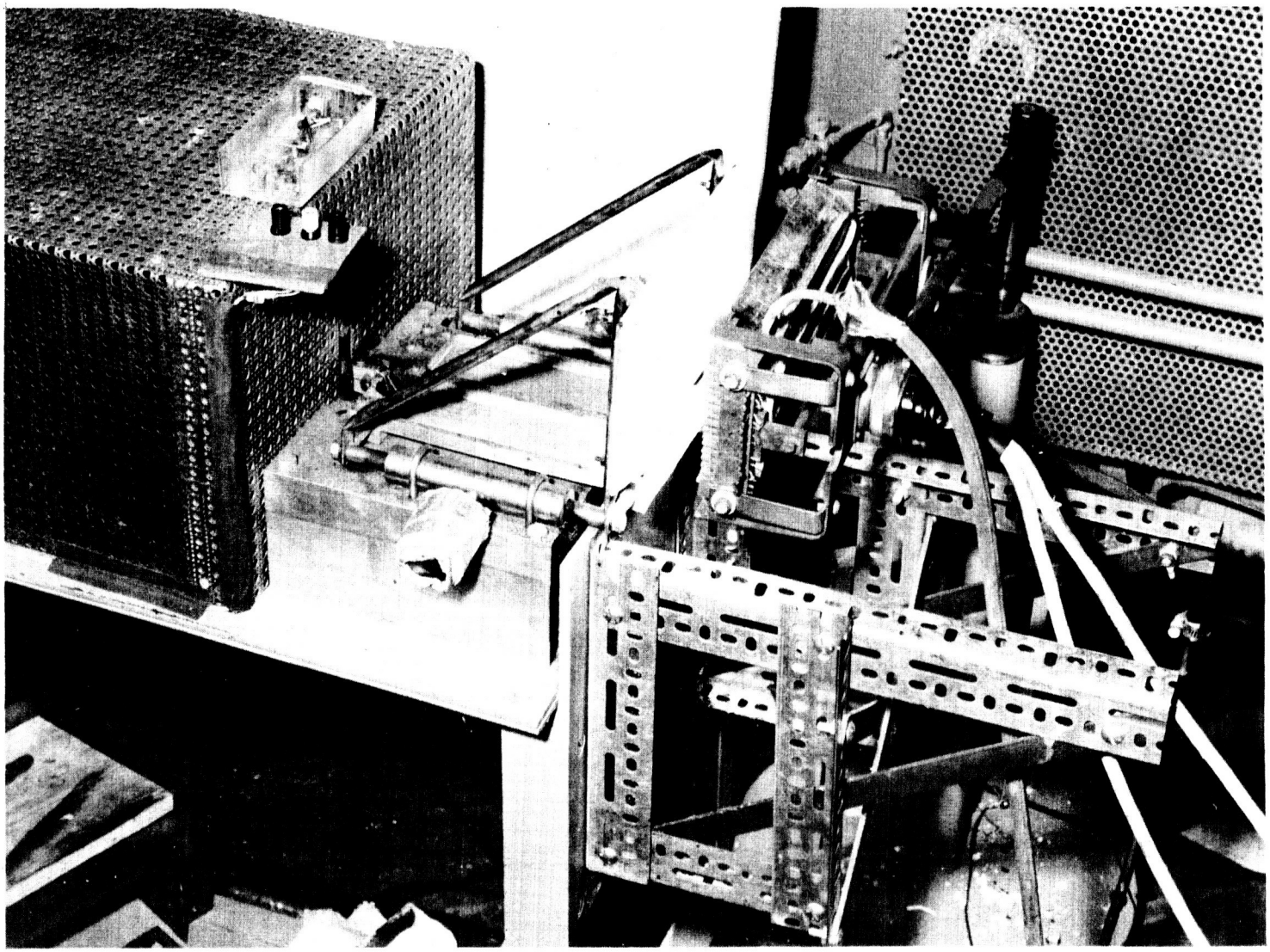

Figure 6 - Radiant Heating Apparatus With M-31 Specimen in Test Position

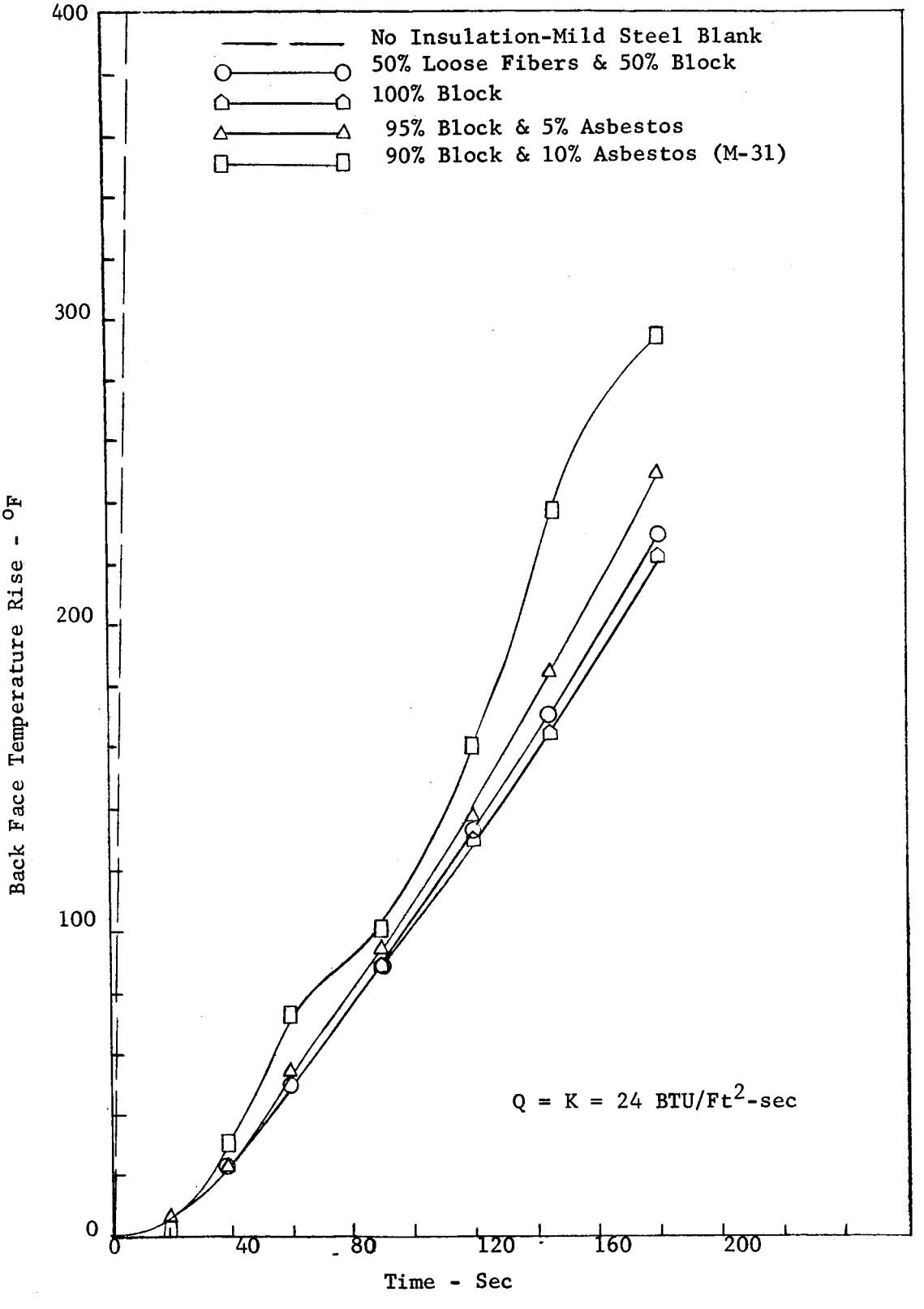


Figure 7 - Effect of Composition on the Temperature Rise of "Tipersul" Ceramic Coatings

MTP-P&VE-M-62-14

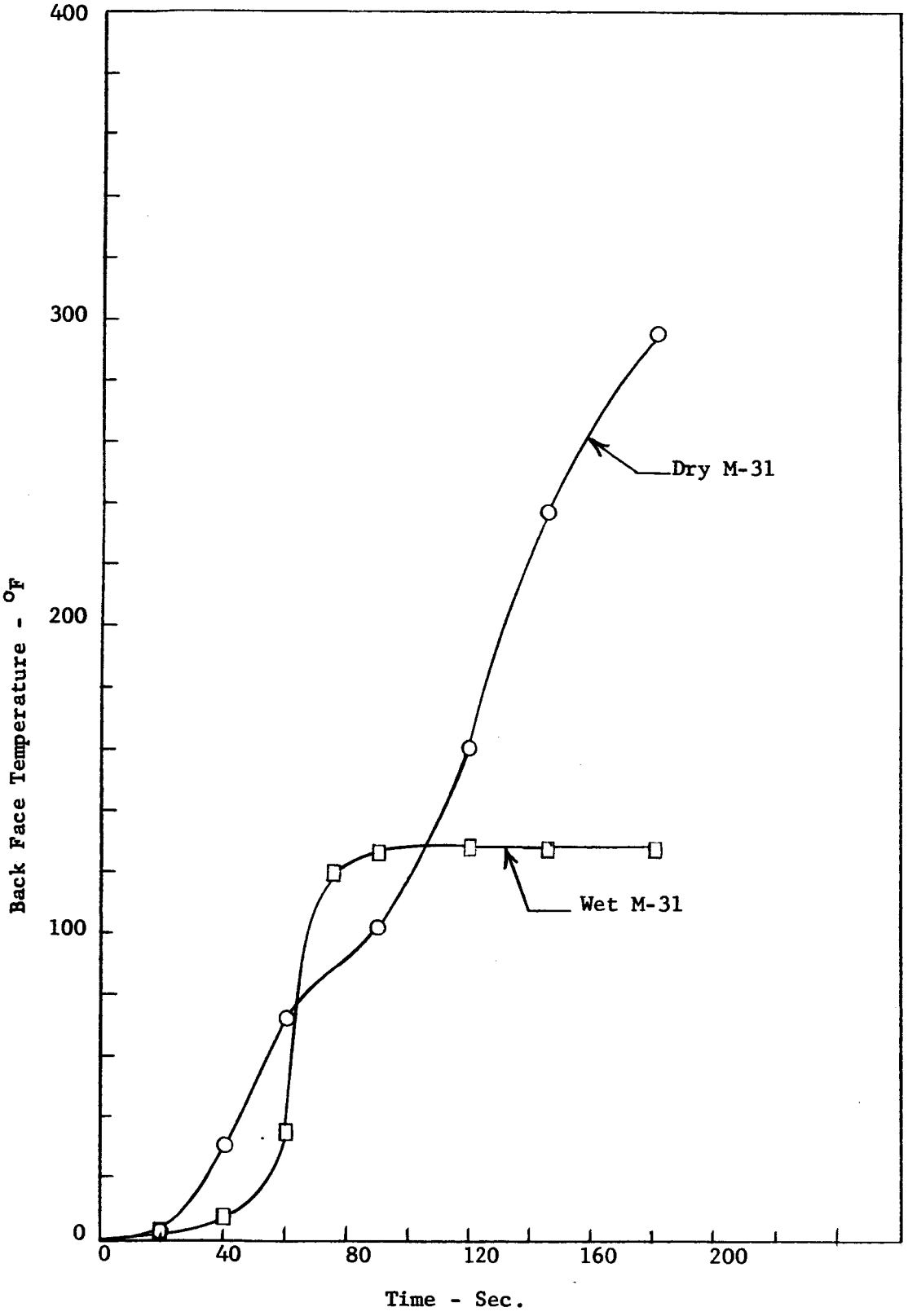


Figure 8 - Effect of Water on the Temperature Rise of M-31

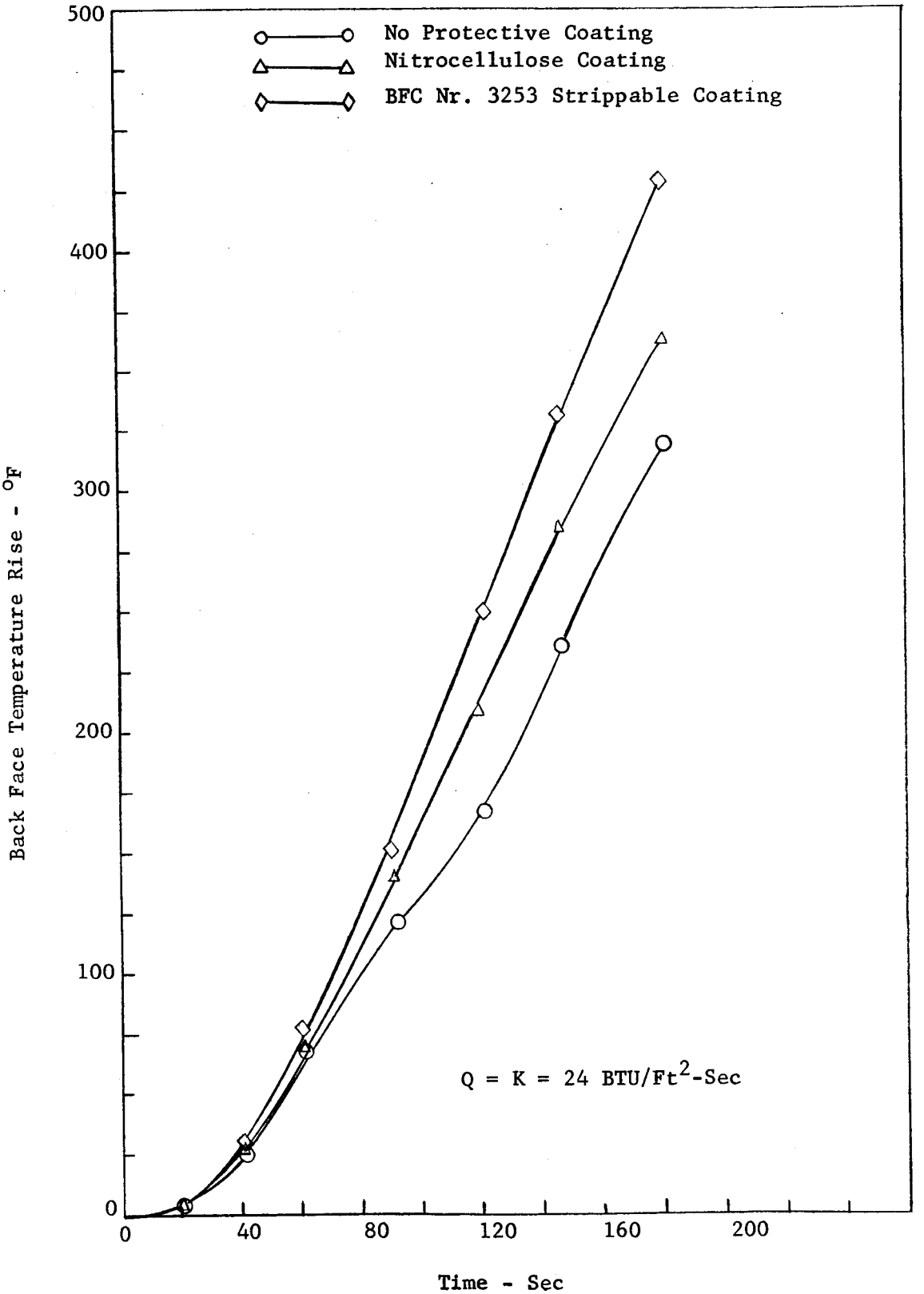


Figure 9 - Effect of Protective Coatings on the Temperature Rise of M-31

APPROVAL MTP-P&VE-M-62-14

DEVELOPMENT OF A HIGHLY REFLECTIVE UNFIRED CERAMIC THERMAL INSULATION

By Vaughn F. Seitzinger

The information in this report has been reviewed for security classification. Review of any information concerning Department of Defense or Atomic Energy Commission programs has been made by the MSFC Security Classification Officer. This report, in its entirety, has been determined to be unclassified.

J. E. Kingsbury
Chief, Engineering Research Section

W. R. Lucas
Chief, Engineering Materials Branch

W. A. Mrazek
Director, Propulsion and Vehicle Engineering Division

DISTRIBUTION

| | |
|---|---|
| M-P&VE-DIR | Dr. Mrazek |
| M-P&VE-DIR | Mr. Hellebrand |
| M-P&VE-F | Mr. Goerner |
| M-P&VE-E | Mr. Schulze |
| M-P&VE-S | Mr. Kroll |
| M-P&VE-SB | Mr. Verble |
| M-P&VE-SA | Mr. Blumrich |
| M-P&VE-P | Mr. Paul |
| M-P&VE-PH | Mr. Connell |
| M-P&VE-PT | Mr. Wood |
| M-TEST-DIR | Mr. Heimburg |
| M-ME-DIR | Mr. Kuers |
| M-QUAL-DIR | Mr. Grau |
| M-P&VE-M | (5) |
| M-P&VE-ADMP | Mr. Hofues |
| M-MS-IP | Mr. Remer |
| M-P&VE-ME | (10) |
| M-MS-IPL | (8) |
| M-MS-H | Mr. Akens |
| M-HME-P | |

Scientific & Tech. Information Facility
Attn: NASA Representatives
(S-AK/RKT)
P. O. Box 5700
Bethesda, Maryland